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13. ABSTRACT (Maximum 200 words)
The original aim of this work was to develop an ultrashort pulse soft x-ray light source. In the course of exploring laser produced plasmas and high order harmonic generation sources this work uncovered a wealth of new physics in the realm of high intensity laser-atom photoionization. The study of the absorption of many photons leading to ionization is of vital importance to understanding novel light sources. This project discovered and exploited a new restricted geometry measurement technique and methodology that has allowed a more direct comparison of experimental data with theory. During the last year the project produced several publications on using restricted geometry detection and uncovered new photoelectron features in kinetic energy spectrum measured from photoionization with high intensity laser pulses resulting from the absorption of over 30 photons by a single atom. These measurements have raised new questions concerning the role of doubly excited states and electron-electron correlations in high intensity photophysics.

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DEVELOPMENT OF AN ULTRASHORT PULSE SOFT X-RAY LIGHT SOURCE

FINAL REPORT

LINN D. VAN WOERKOM

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U.S. ARMY RESEARCH OFFICE

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THE OHIO STATE UNIVERSITY

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FINAL PROGRESS REPORT

ARO PROPOSAL #33789-PH

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"DEVELOPMENT OF AN ULTRASHORT PULSE SOFT X-RAY LIGHT SOURCE"

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Statement of the Problem Studied

The problem studied dealt with developing a light source of ultrashort pulse duration in the vacuum ultraviolet (VUV) to x-ray regions of the electromagnetic spectrum utilizing the interaction of high intensity, ultrashort pulse laser light.

Important Results

This report describes work performed on developing an ultrashort pulse soft x-ray light source using the interaction of high intensity ultrashort pulse laser light with matter. The time period covered for this project was 15 July 95 – 14 July 98. In order to develop a useful laboratory light source it is necessary to understand the underlying physics of the interaction of light and matter. The three main areas of research covered during the reporting period were:

- Laser produced plasma light source
- High harmonic generation light source
- High intensity multiphoton and above threshold ionization

Laser Produced Plasmas

The first project involved the pulse duration measurements of laser produced plasma (LPP) soft x-rays. The basic idea was to use a pump-probe geometry to gate the transmission of x-rays through an absorbing medium. Soft x-rays produced by the interaction of 800 nm, 140 fs pulses with brass targets were imaged from the plasma to an interaction region located at the exit nozzle of a pulsed gas jet containing krypton. The soft x-rays excited a transition in krypton from a 4d inner-shell level to a 5p Rydberg state at about 91.5 eV. The subsequent absorption of the x-rays was monitored using a custom built 1.5 m radius Rowland circle grazing incidence spectrometer coupled to a custom microchannel plate intensified phosphor screen. Under normal conditions (i.e. no probe pulse), a clean absorption feature at 91.5 eV was observed. By introducing an appropriate optical delay it was possible to also irradiate the krypton gas jet with a portion of the original 800 nm laser beam. The presence of the intense laser light created significant ionization and thus destroyed the population of neutral atoms necessary for absorption of the x-rays. This resulted in an increase in transmitted x-rays and a loss of the absorption profile. The delay was changed and the temporal evolution of the convolution of the soft x-rays and the driving laser was mapped out.

Unfortunately, the duration measurements indicated soft x-ray durations on the order of tens to hundreds of picoseconds. Similar results were found at the University of Michigan in Donald Umstadter's group. The results were largely unpublishable except for some very nice work on imaging.[1] The problem lies in the fact that at very high intensities ($>10^{15}$ W/cm^2) the system is overdriven in so far as the plasma becomes so hot that energy transport actually slows down the cooling times and creates long emission times. As mentioned in the original proposal for this work, our approach is to find a usable source and not become entangled in the plasma physics of the LPP source.

As mentioned above, the duration of the soft x-rays emitted from solid targets upon irradiation with intense laser light is well above one picosecond. As a final experiment in using laser produced plasmas we measured high resolution x-ray spectra from about 4 - 40 nm for various solid targets. In particular we were interested in using transparent targets in order to transmit lower intensity light without creating a plasma. Our results were published in Physical Review E as a Brief Report.[2] In short, we measured fairly narrow spectral features over a wide range indicating that the radiation was emitted from a longer scale length plasma (and hence lower density). The experiment resulted in beautiful data that will serve as database information for future research. Upon completion of this experiment we dismantled the laser produced plasma apparatus.

High Harmonic Generation

Still in search of a usable laboratory light source, we shifted direction away from LPP's in favor of high harmonic generation (HHG). This process has the distinct advantage that the harmonic radiation must be ultrashort in duration since it is created by the nonresonant, nonlinear polarization of the medium driven by the ultrashort laser pulse. However, the total number of photons emitted into a given harmonic is small, indicating that a usable light source must address the issues of efficiency.

The second project was the development of a high harmonic generation (HHG) apparatus in which to study both the production and application of short wavelength ultrashort duration pulses. To this end we designed and built several differentially pumped vacuum chambers in order to transport the harmonic radiation from the moderate pressure regions near the gas jet to high vacuum regions for detection. Detection was accomplished with an f/4.5 normal incidence vacuum ultraviolet (VUV) spectrometer. Experiments were concurrently being designed to separate out the harmonics with the spectrometer and re-image single harmonics into a time-of-flight (TOF) spectrometer for ion and photoelectron analysis.

Atom/Molecule	Ip (eV)	Static average polarizability (1e-24 cm3)
C4H10	10.57	8.2
C3H8	10.95	6.29
C2H6	11.52	4.47
N2	15.58	1.74
H2	15.43	0.8
SF6	15.33	6.54
Ar	15.76	1.64
Kr	14	2.48
Xe	12.13	4.04

Table 1. Ionization potential and polarizability for the atoms and molecules studied.

Furthermore, a variety of gases were studied in order to find more efficient harmonic sources. Two main experiments were performed using the HHG source. First, a study of harmonic efficiency using different molecular gases was performed. In particular, the goal was to study trends relating harmonic efficiency with the static polarizability of the target species. The gases studied and the corresponding ionization potentials and polarizabilities are given in Table 1.

The main results showed that in the tight focusing limit the efficiency for producing harmonics in the range of 7th-15th harmonic followed the polarizability. More polarizable molecules such as propane produced nearly as much harmonic signal as xenon. It remains true, however, that xenon is the most efficient HHG target gas. Figure 1 shows the main result of the experiment. Key areas of research that remain include understanding the fragmentation properties of molecules in intense fields. It is not known if the harmonics are produced from the neutral parent molecule, neutral fragments, or ionized fragments.

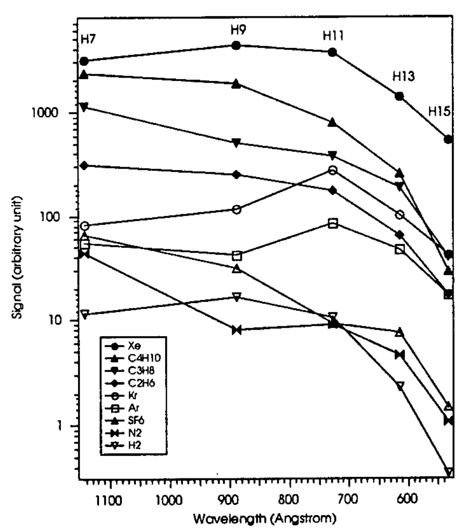

Fig. 1 – HHG with various gases

The second set of experiments involving the HHG source centered around using the harmonics as a source for photoionization of gases. In particular, the goal of our first set of experiments was to look for nonlinear optical effects with high harmonics. Using our f/4.5 normal incidence VUV spectrometer as both a spectral filter and re-imaging optic, we imaged given harmonics into the interaction region of a time-of-flight ion/electron spectrometer. By tuning to particular harmonics, we looked for two-photon ionization with a high harmonics.

The initial measurements looked promising but proved to be due to contamination from higher order diffraction of higher harmonics. In addition, upon ray-tracing the optical system we found that the focal spot of the harmonic was large and astigmatic. This lead to an overall decrease of the intensity and hence killed any chances of seeing two-photon processes. Improvements are being made to the apparatus in the hopes of using the harmonics for time-resolved studies of strong field ionization.

Multiphoton Ionization and Above Threshold Ionization

Finally, the third project area involved the study of high laser intensity multiphoton ionization. The original course of events was aimed at developing a high efficiency photoelectron time-of-flight (TOF) setup. One of my graduate students, Peter Hansch, manufactured an ellipsoidal mirror TOF spectrometer based on reflecting electrons from grid optics.[3] The goal was to find a way to keep high resolution given an extended photoelectron source. Since any x-ray source scheme involves re-imaging the light, and since most processes are linear in the intensity for short wavelengths, it was necessary to develop a strategy that would keep high resolution for large source size.

In the course of developing this detector we encountered a tremendous wealth of new physics. The detector was tested on our kHz laser system for calibration. With the tremendous data collection rate afforded by the 1 kHz repetition rate, it was possible to obtain an incredibly large signal to noise ratio. We pursued high intensity photoionization of the noble gases and found a preponderance of electrons with kinetic energies as high as 500 eV. Only two other groups in the world had observed these electrons (DiMauro at Brookhaven National Laboratory, and Paulus at the Max Planck Institute).[4-6] We began a study of this phenomenon since it involved coupling a large (~100 eV or more) amount of energy into a single atom. Such processes are important for understanding the production of x-rays with high harmonic generation. While this portion of the work represents a slight deviation from the original mission of developing a light source, we have uncovered new science in the ensuing time. Below are listed the most important results of this research effort.

1. Intensity Selective Scanning

One of the major limitations to deciphering intensity dependent phenomena is spatial averaging due to the continuous distribution of intensities in a Gaussian focus. We developed a new technique which allows us to view very small portions of the focal region, thereby controlling both the intensity and the volume weighting.[7] Instead of exposing the entire ionization region to the time-of-flight detector, a pinhole at the front of the flight tube drastically reduces the relevant ionization volume which is in the line-of-sight of the detector. By moving the focusing lens, the ionization region scans across the pinhole allowing intensity-selective observation of MPI in a field-free region *without* changing the characteristics of the focused laser beam, while seeing a significant signal gain. As the focused laser beam diverges from its minimum beam waist the volume occupied by lower intensities increases rapidly, producing large count rates for a wide variety of intensities. Typical high order MPI (requiring greater than 5 photons to ionize) spans the range from 10^{12}-10^{14} W/cm^2, beyond which tunneling processes become non-negligible. The ISS method allows observation of low intensity MPI by selecting smaller local intensities with the pinhole along the z-direction, while maintaining the same absolute I_0. The pinhole of diameter Δz allows only a small fraction of the total focal volume to be seen by the detector. However, the radial expansion of low intensity contours with increasing z coordinate causes a significant increase in low intensity volume so that the overall signal maintains an excellent signal to noise ratio. The ISS approach differs from traditional TOF photoelectron spectroscopy by blocking out the otherwise dominating high intensity signal. The well known problem of spatially averaging over a large intensity range is reduced; instead, observation of a radial one-dimensional intensity distribution is now possible, providing much more reliable data for comparison with theoretical MPI models.

2. Removing Spatial Averaging

We used this technique for measuring both electron kinetic energy spectra as well as ion charge state distributions. Traditionally, for a given ion charge state the ion yield after saturation continues to rise as the intensity increases due to the growth of the entire focal volume. The true ionization rates (intensity-dependent, not volume-dependent) can only be extracted by deconvolving the complicated Gaussian volume. Despite the low ionization rates at low intensities, the volume increase in a Gaussian focal volume causes a significant number of ions to be produced far away from the center of the beam. In fact we recently showed that a non-negligible fraction of Xenon ions can be generated as far as several Rayleigh ranges away from the minimum beam waist.[8] In addition, since the intensity distribution is reduced to a one-dimensional slice we deconvolved the ionization probability from the signal.[9] Thus, we now have the ability to produce ion or energy specific electron yields as a function of laser intensity without any spatial averaging. This is particularly important since we can choose an electron energy peak and follow its evolution as a function of intensity to clarify the role of specific resonances.

3. Transient Resonances

We studied transient resonances in xenon with high resolution and high precision. This work is an extension and verification of the transient resonance model that has been the standard in the field since 1987.[10] The idea is that an initially nonresonant laser field produces multiphoton resonantly enhanced ionization due to the a.c. Stark shift of high lying Rydberg states. Many groups studied the lowest multiphoton resonances in the noble gases over the last decade. Our work, however, concentrated on the next higher multiphoton resonances and illustrated with clarity the physics using kilohertz lasers and the ISS technique.[11] For these experiments the absolute peak intensity $I_0=6.4\times10^{13}$ W/cm^2 at the minimum beam waist was held fixed while spectra over the range 5×10^{12} - 6.4×10^{13} W/cm^2 were recorded via Intensity-Selective-Scanning (ISS) as described above.

We modeled the evolution of the 8- and 9-photon resonances using multiphoton Landau-Zener theory. The ground state is dressed in energy with either 8 or 9 photons to create an avoided crossing with the time dependent energy levels of the Rydberg states. Saturation effects due to rising and falling edge ionization are also included and a full three-dimensional spatial integration is performed using the geometry of a volume slice for each spectrum.

4. Hot Electrons

The most important result of our research involves new physics in ATI. As mentioned above, we measured photoelectron spectra for noble gases. Figure 2 shows data illustrating the high resolution and dynamic range of our

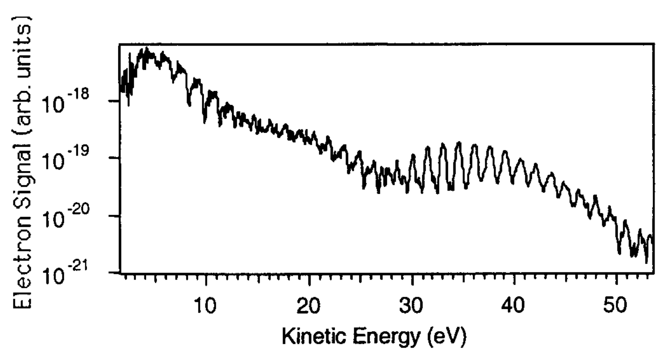

FIG. 2 - Xenon

photoelectron measurements in xenon. Of particular interest is the prominence of the hot electron tail and bump near 35 eV. We are able to follow the evolution of the photoelectron spectrum from low kinetic energies and low laser intensity to high KE and high intensity. These spectra are rich in structure that is not well understood at this time. Recently we observed new sub-structure in the hot electron portion of the xenon spectrum.[12] Current theories are unable to predict the sub-structure although recent calculations raise the issue of doubly excited states. It appears that a resonant process is responsible with a very restrictive intensity dependence.

Due to the high kinetic energies (15-40 eV) and the sensitive intensity dependence, it is difficult to explore this region theoretically. The kinetic energy range shown in Fig. 2 includes many excited states of the xenon ion with series of neutral autoionizing levels. The role of these states with more than one excited electron must be determined. Thus, more careful experiments must be performed in order to elucidate the physics of these features.

We also performed similar experiments on argon which show that some of the high kinetic energy structure follows the intensity dependence of the neutral transient Rydberg resonances as shown in figure 3. Due to our fast electronics we achieve a resolution of 30 meV beyond 30 eV. By carefully following the evolution of the transient resonances with laser intensity at low kinetic energies we can absolutely calibrate the energy scale to better than 50 meV over the range of energies from 0 - 50 eV. The narrow features within each ATI order near 21 eV are real and in fact follow the intensity dependence of the low energy Rydberg features. This is evident in figure 3. The insets show the Rydberg structure for low KE (top inset) and the corresponding ATI orders after absorbing 15 additional photons (bottom inset). Muller's recent calculations also reproduce this behavior without going beyond the SAE model.[13] This is a very interesting energy region that has yet to be fully understood.

It is interesting to notice that the enhancement in photoelectron signal

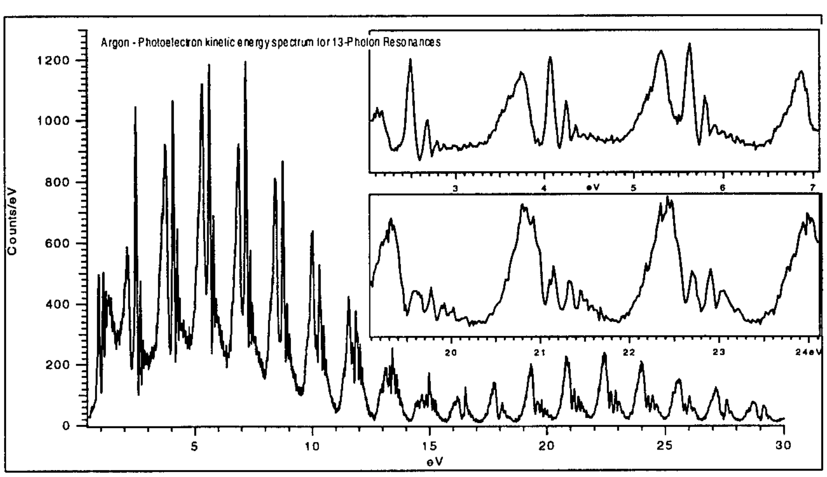

Fig. 3 – High Order ATI in Argon

begins when the photoelectrons are emitted with kinetic energy close to the ionic excited states. Many neutral autoionizing states exist that converge on the excited ion levels. Thus, one hypothesis is that the very high order ATI enhancement is due to ionization into a structured continuum. We observed a similar alignment of the excited ionic states with the ATI enhancements for xenon, krypton and argon. The enhancements appear at slightly different kinetic energies for each atom and the sub-structure varies from single peaks at high kinetic energy for xenon to narrow Rydberg structure in argon. A second interpretation is that the modulation in the electron spectrum is due to the interference of the tunneling and multiphoton components of ionization. This idea is attractive due to the intermediate intensity at which these experiments have been performed. The Keldysh parameter is about 1.5 which indicates that multiphoton effects should dominate, but tunneling processes are not necessarily negligible. More detailed experiments are needed that measure the angular distributions and electron spectra for a variety of atoms in order to unravel the role of specific angular momentum states. In particular, direct comparisons with argon are now possible.

Conclusion

In conclusion, this research program explored the generation and application of short pulse radiation from the vacuum-ultraviolet to the soft x-ray regions of the spectrum. The efforts were not successful in producing a useful sub-picosecond soft x-ray source from laser produced plasmas. Experiments using high harmonic generation illustrated the usefulness of HHG as a light source but failed to show nonlinear behavior in photoionization. Finally, a very productive program in multiphoton ionization and above threshold ionization emerged utilizing kilohertz laser sources and unique applications of time-of-flight spectroscopy.

Publications resulting from this research

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Scientific Personnel

Professor Linn D. Van Woerkom, PI

James Norby, Ph.D. in Physics 1995

Peter Hansch, Ph.D. in Chemical Physics 1997

Son Evans, Ph.D. in Physics 1997

Mark Walker, graduate student

Report of Inventions

No inventions

Technology Transfer

No technology transfer

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